MEASUREMENTS OF BLACK CARBON IN CALIFORNIA SNOW AND RAIN

PIER FINAL PROJECT REPORT

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Preface

The California Energy Commission's Public Interest Energy Research (PIER) Program supports public interest energy research and development that will help improve the quality of life in California by bringing environmentally safe, affordable, and reliable energy services and products to the marketplace.

The PIER Program conducts public interest research, development, and demonstration (RD&D) projects to benefit California.

The PIER Program strives to conduct the most promising public interest energy research by partnering with RD&D entities, including individuals, businesses, utilities, and public or private research institutions.

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- Renewable Energy Technologies

- Transportation

Measurements of Black Carbon in California Snow and Rain is the final report for the Evaluating Past and Improving Present and Future Measurements of Black Carbon Particles in the Atmosphere project (contract number 500-02-004, work authorization number MR-06-01B) conducted by Scripps Institution of Oceanography.

For more information on the PIER Program, please visit the Energy Commission's website www.energy.ca.gov/pier/ or contract the Energy Commission at (916) 654-5164.

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Abstract

Black carbon, a main component of combustion-generated soot, is a strong absorber of solar radiation and a major contributor to climate change. Black carbon contributes to global warming by direct solar heating of the atmosphere and also by reducing the reflectivity (albedo) of ice and snow through deposition. Contamination occurs largely by wet deposition, so it is important to know the amount of black carbon in rain and snowfall. This study undertook the first such measurements in California. Automated rain and snow collectors were deployed February–May 2006. The collected precipitation was analyzed for black carbon as well as for iron, calcium, and potassium. The average black carbon concentration in coastal rain was 5.7 ± 3.5 nanograms/gram. The average black carbon concentration in mountain snow was 5.0 ± 2.3 nanograms/gram at the remote site and 6.9 ± 3.1 nanograms/gram at the polluted site. The data reveal that snowfall removes almost all atmospheric black carbon, which is deposited over the snowpack. Comparisons with precipitation amount per day and ambient concentrations in the air indicate that concentrations of black carbon and other aerosol species are primarily dominated by emissions originating in California. Calculated air mass back trajectories also indicate strong local/regional sources for the aerosol, including black carbon, in precipitation. Evidence of long-range transport was measured for isolated events but additional field research is needed to undertake seasonal deposition of black carbon over the Sierra/Cascades and its impact on snowmelt rates.

Keywords: Black carbon (BC), precipitation, snow melt, climate change, aerosol light-absorption, particulate matter

Executive Summary

Introduction

Soot particles are emitted by combustion of fossil fuels (such as diesel), biofuels (such as wood fires), and biomass (during forest fires). Soot contains many chemical species (elemental carbon and hundreds of organic acids) that both scatter and absorb solar radiation in the atmosphere. The particles in soot that absorb solar radiation are referred to as *black carbon.*

Black carbon is an important source of global warming and a major contributor to the melting of sea ice, snow, and glaciers. It is removed from the atmosphere mainly through wash-out in rainfall and snowfall. Black carbon particles deposited by snowfall over snow (or ice) "darken" the snow (or ice) and increase the amount of radiation absorbed by snow (or ice), which increases snow and ice melt rates. Enhanced melting exposes the underlying darker surface (land or sea), which enhances solar absorption by about 50 percent or more and contributes to a substantial surface warming. This same mechanism can contribute to snowpack melting in the Sierra and thus affect California's freshwater availability.

Until now, studies on the effects of black carbon on snow and ice have focused mainly in the Arctic. Very few observational studies, if any, have been conducted regarding black carbon's effects on California's snowpack. Two sources contribute to black carbon pollution in California high country. Local and regional black carbon emissions from California's industrial and agricultural centers are transported by prevailing westerly winds to the Sierra Nevada/Cascades and washed out in falling snow. The second source of black carbon is from long-range transport of pollution from East Asia across the Pacific Ocean. Research by the authors has shown that significant quantities of pollution are transported from Asia and Europe to the West Coast every year. These transported pollutants affect high-elevation sites more frequently than low-lying regions.

Purpose

This project sought to determine the black carbon concentration in:

- Snow in pristine and polluted areas
- Coastal rain.

The work was funded as part of the Suppression of Precipitation Experiment (SUPRECIP-2), which studied how aerosols affect clouds and precipitation in the Sierra Nevada.

Project Objectives

This project's objectives were as follows:

- Given the finding of substantial black carbon deposition in snow, collect data over longer periods and at more stations distributed over elevated sites in California.
- Analyze snow and rainwater at three locations in Northern California to determine black carbon concentration in the precipitation.

- Use meteorological wind fields and ambient aerosol measurements to obtain information on the source of the black carbon in the rain.
- Place the observations in context with previous model results to assess possible effects on California's mountain snowpack.

Project Outcomes and Conclusions

This study is the first experimental study of black carbon deposition on California snowpacks. The research team found black carbon concentrations in snow in both pristine and polluted mountain areas and in coastal rain. Between February 15 and May 15, 2006, 15 snow samples were collected from two high-elevation sites (in the Sierra Nevada and southern Cascades) and analyzed for black carbon concentration. Fourteen rain samples were collected at the coast and analyzed for black carbon to determine its concentration in precipitation as it reaches North America. The average black carbon concentrations in rain at Trinidad, California (Humboldt County), and in snow at Central Sierra Snow Lab and Lassen Volcanic National Park were, respectively, 5.7 ± 3.5 nanograms per gram (ng/g), 6.9 ± 3.1 nanograms per gram, and 5.0 ± 2.3 nanograms per gram. These values are typical of concentrations found in snow in urban environments. Comparison with ambient atmospheric concentrations of black carbon reveals that most of the black carbon is incorporated into the falling snow below the cloud, effectively removing the black carbon particles from the atmosphere and depositing them into the snowpack, which potentially could have drastic effects on water availability in California. During the period of observations, local and regional sources were the major contributors to the observed black carbon.

Recommendations

Some vital pieces of information about black carbon in snowpack remain unknown, and this complicates the conclusions this report can draw. For example, snow-melt rates in April, when the rainy season begins, need to be determined. Also, the movement of black carbon through snowpack during snow melts also is not well understood. Because of these unknown factors, a more detailed study on the seasonal deposition of black carbon over the Sierra/Cascades and its impact on snowpack melt rates should be undertaken.

Benefits to California

Mountain snowpack is an important reservoir for freshwater in California. Studies have shown that the onset of spring melt has been occurring earlier in the year, coincident with the spring rainy season. This study better characterizes how a faster melt rate due to black carbon contaminants in the snow may affect the situation. The combined result could produce more flooding earlier in the summer and lack of available water in the late summer and early autumn.

Technical Summary

Introduction

Black carbon aerosols scatter and absorb solar radiation. Most studies show that the net effect is a positive forcing (more incoming energy; therefore, a net warming effect) on the climate. In addition to *direct* radiative forcing, black carbon also affects the radiation budget through its influence on snow surfaces. Black carbon particles in snow or ice increase the amount of radiation absorbed at the surface and lower the albedo, or reflectivity, of the snow. Not only does this exert a positive forcing on the radiation budget, it could also lead to an increased snow and ice melt rate. Enhanced melting not only decreases the planetary albedo by reducing the surface area of highly reflective snow and ice, but may also impact freshwater resources for regions like California that rely on mountain snowpacks for water in dry summer months.

Several field studies, conducted within the last decade have shown that trans-Pacific transport of pollution, including black carbon, from Asia to North America can be significant, especially in the late winter and early spring (*Bertschi and Jaffe,* 2005; *Bey et al.,* 2001; *de Gouw, et al.* 2004; *Goldstein et al.,* 2004; *Hadley et al.,* 2007; *Heald et al.,* 2006; *Liu et al.,* 2003; *Park et al.,* 2005; *Parrish et al.,* 2004). Not only does the timing of this enhanced transport occur at the same time as the onset of spring melt, it also impacts high-elevation sites with greater significance (*Hadley et al.,* 2007; *VanCuren et al.,* 2005). A recent long-range aircraft experiment using the NCAR-GULF V (*Stith and Ramanathan,* 2007, see http://borneo.ucsd.edu/SIO-PACDEX.html) tracked soot-dust plumes from the western Pacific to North America. Therefore, the black carbon observed in California precipitation is likely a combination of both local and transported sources, especially in March and early April.

Recent modeling studies have predicted that snow surface albedo is reduced by approximately 0.2 percent per nanogram of black carbon per gram of equivalent snow water. Reduction in albedo is extremely relevant to snowpack melt, as more incoming solar radiation will be absorbed at the surface. Solar radiation is the primary driver of melting when ambient temperatures reach 0°C, as can be observed when comparing snow cover on north- and south-facing mountain slopes during the spring melt.

Snowpack in the Sierra Nevada and Cascade ranges is a crucial freshwater reservoir for California's massive agricultural production, as well as for densely populated urban and suburban centers. Measurements of actual black carbon concentration in the snow are necessary to place these modeled effects into a relevant context for California.

Purpose

This project determined the black carbon concentration in snow in pristine and polluted areas and in coastal rain. These observations are placed in context with model results to determine the effects of black carbon on the California mountain snowpack. Additional aerosol and meteorological measurements provide information on likely sources for the observed black carbon in coastal and mountain precipitation.

Project Objectives

The objectives of this project are as follows:

- Given the finding of substantial black carbon deposition in snow, collect data over longer periods and at more stations distributed across elevated sites in California.

- Analyze snow and rainwater at three separate locations in Northern California to determine black carbon concentration in the precipitation.

- Use meteorological wind fields and ambient aerosol measurements to determine the source of the black carbon in the rain.

- Place the observations in context with previous model results to assess possible impacts on California mountain snowpack.

Project Outcomes

Between February 15 and May 15, 2006, 21 precipitation samples, of which 14 were large enough to analyze for black carbon, were collected at Trinidad Head (THD). At Mt. Lassen Volcanic National Park (LAVO), 17 samples were collected, and 7 of these were of sufficient size to analyze for black carbon. Seventeen samples were also collected at Central Sierra Snow Laboratory (CSSL), of which 8 were suitable for black carbon analysis. The smaller samples at CSSL and LAVO were analyzed for iron, calcium, and potassium for comparison with ambient atmospheric concentrations available at these sites. The average black carbon concentrations in rain at THD, and in snow at CSSL and LAVO, were, respectively, 5.7 + 3.5 nanograms per gram (ng/g), 6.9 ± 3.1 ng/g, and 5.0 ± 2.3 ng/g. Similar measurements made by Chylek et al. (1999) near Halifax, Nova Scotia, showed that black carbon concentration in rural Nova Scotia averaged 1.7 ± 0.83 ng/g, while those in urban Halifax averaged 11 ± 7.7 ng/g.

The experimental details and results have been presented in three conferences:

Hadley, O. L., C. E. Corrigan, V. Ramanathan, T. W. Kirchstetter, and S. Cliff. "Black Carbon in Rain and Snow in the Sierra Nevada Mountains." Oral presentation at the AGU (American Geophysics Union) 2007 fall meeting, San Francisco, California.

Hadley, O. L., C. E. Corrigan, V. Ramanathan, T. W. Kirchstetter, and J. Aguiar. "Application of a New Thermal-Optical Evolved Gas Analysis Method to Quantify Black Carbon in Precipitation." Poster presentation at the AAAR (American Association for Aerosol Research) 26th Annual Conference, 2007, Reno, Nevada.

Hadley, O. L., C. E. Corrigan, V. Ramanathan, T. W. Kirchstetter, and J. Aguiar. "Black Carbon in Rain and Snow in the Sierra Nevada Mountains." Oral presentation at the California Energy Commission 4th Annual Research Conference on Climate Change, 2007, Sacramento, California.

This study's results are also being prepared as two journal papers. The first will document the experimental techniques and the data. The second paper will present the results for black

carbon content of snow along with an impact estimate on the radiative forcing of the snowpack in California.

Conclusions

Aerosol analysis and air-mass back-trajectory calculations for CSSL both point to a strong local influence (within 100 kilometers). Analysis for LAVO also shows predominantly local influences but includes isolated incidents of trans-Pacific transport of pollutants. High correlation between the surface air concentration of black carbon just before a precipitation event and the total amount of black carbon deposited during the event indicate that black carbon is incorporated into snow via below-cloud scavenging. Further, the data show that nearly all of the black carbon in the boundary layer is removed by the falling snow. At THD, the two highest observed concentrations correspond to days where air mass back trajectories indicate direct trans-Pacific transport. Back-trajectory calculations for the other sample days show that the air masses spent more time over the ocean and, furthermore, show strong cyclonic rotation over the Pacific Ocean, representing strong precipitation events prior to arrival at THD.

Based on the results of several modeling studies, the black carbon concentration found in the snow at the mountain sites would theoretically result in an initial reduction in surface albedo[1] for fresh snow between 0.35 percent and 1 percent, for visible wavelengths and assuming snow crystals with an effective radius of 100 microns (*Flanner et al.*, 2007; *Hansen and Nazarenko*, 2004; *Jacobson* 2004). All model results show that for larger snow crystals and aged snow, the effectiveness of black carbon at reducing albedo and absorbing solar radiation increases dramatically. This could lead to a strong positive feedback in early springtime snow melt. A major uncertainty is that it is unknown how black carbon distributes itself throughout the snowpack during melt. Many models assume that as snow melts, its black carbon is left behind in the surface layers, further amplifying its effect, although it is possible that much of the black carbon is flushed through the snow with the melt water.

Recommendations

Additional research is needed in the following areas:

- Field studies to determine the vertical redistribution of black carbon throughout the snowpack during the melt season.
- Direct measurements of snow albedo along with the effective radius of the snow crystal size, for validation the theoretical relationship between albedo and black carbon concentration.
- Incorporation of an analysis method requiring small volumes of water with automated rain samplers, which would allow high-temporal-resolution measurements of black

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[1] The ratio, expressed as a percentage, of the amount of electromagnetic radiation reflected by the earth's surface to the amount incident upon it. The value varies with wavelength and with the surface composition.

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carbon concentration in precipitation as it changes throughout an event. These measurements could help separate "washed out" aerosols from those in the original cloud-condensed nuclei, to help determine the precipitation removal efficiency of black carbon.

Benefits to California

Available freshwater during the summer months in California is a major concern for both urban population centers and agriculture, which represents a significant fraction of the California economy. Mountain snowpack is an important reservoir for California's freshwater. Studies have shown that the onset of spring melt has been occurring earlier in the year, coincident with the spring rainy season. This phenomenon, combined with a faster melt rate due to black carbon contaminants in the snow, means the melt water will have nowhere to go when it reaches already full reservoirs–which could result in more flooding earlier in the summer and lack of available water in the late summer and early autumn.

1.0 Introduction

Black carbon (BC) or soot particles are unique in the atmosphere, both as efficient absorbers of radiation at all wavelengths and as hydrophobic particles not as readily removed by precipitation as other aerosols. Unlike most aerosols, which primarily scatter solar radiation, enhance planetary albedo, and cool the planet, BC particles absorb incoming solar radiation and warm the atmosphere (*Jacobson*, 2001; *Ramanathan, et al.*, 2005; *Satheesh and Ramanathan*, 2000). Due to the relative inefficiency of wet removal for BC particles (*Zuberi, et al.*, 2005), a longer atmospheric residence time increases the duration of positive forcing on the climate. Aged BC particles that are incorporated into cloud drops may heat the cloud, evaporate the droplets, and shorten cloud lifetime (*Ackerman, et al.*, 2000; *Nenes, et al.*, 2002; *Vogelmann, et al.*, 2001). BC particles not only affect the planetary radiation budget in the atmosphere, but once deposited to ice and snow surfaces, they absorb solar radiation, reduce the albedo, and likely enhance the melt rate of mountain snowpacks, sea ice, and glaciers (*Hansen and Nazarenko*, 2004; *Jacobson*, 2004; *Warren and Wiscombe*, 1980). Enhanced melting not only changes planetary albedo by reducing the surface area of highly reflective snow and ice, but may also impact freshwater resources for regions, like California, that rely on mountain snowpacks for water in dry summer months.

Several field studies conducted within the last decade have shown that trans-Pacific transport of pollution, including BC, from Asia to North America can be significant, especially in the late winter and early spring (*Bertschi and Jaffe*, 2005; *Bey, et al.*, 2001; *de Gouw, et al.*, 2004; *Goldstein, et al.*, 2004; *Hadley, et al.*, 2007; *Heald, et al.*, 2006; *Park, et al.*, 2005; *Parrish, et al.*, 2004). This well documented, enhanced springtime transport of pollution is due to an increase in the number of cold fronts over Inner Mongolia and Taklimakan desert regions, which lift and carry aerosols to about 10 km in altitude; from these altitudes they are transported great distances across the Pacific Ocean by the mid-latitude westerlies (*Liu, et al.*, 2003; *Stohl*, 2001). Not only does the timing of this transport coincide with the onset of spring melt, but it has the greatest impact at high elevations (*Hadley, et al.*, 2007; *VanCuren, et al.*, 2005). Therefore, the BC observed in California precipitation at high elevations and at the coast is likely a combination of both local and transported sources, especially in March and early April.

Recent modeling studies have predicted that snow surface albedo is reduced by approximately 0.2% per nanogram of BC per gram of equivalent snow water (*Jacobson*, 2007). This relationship, however, applies only to fresh snow crystals with an effective radius of 100 μm. As the snow ages, melts, and refreezes, the crystals grow ever larger. This amplifies the effect of the BC inclusions by more than a factor of three. Reduction in albedo is extremely relevant to snowpack melt, as more incoming solar radiation will be absorbed at the surface. Solar radiation is the primary driver of melting when ambient temperatures reach 0°C, as can be observed when comparing snow cover and depth on north- and south-facing mountain slopes during the spring melt. When air temperature falls significantly below freezing, the snow will

remain frozen and a lower albedo will have no effect on melt. Conversely, as air temperature climbs, the melting ascribed to the additional solar energy absorbed by the snow becomes negligible compared with transported sensible heat flux.

Snowpack in the Sierra Nevada and southern Cascade ranges is a crucial freshwater reservoir for California's extensive agricultural production, as well as for densely populated urban and suburban centers. Measurements of actual BC concentration in the snow are necessary to place these modeled effects into a context relevant to the state of California.

To obtain such measurements, this project installed automated precipitation collectors in three California sites for the purpose of collecting and analyzing snow and rain for BC concentration, as well as other aerosol components. Over 55 precipitation samples were collected between late February and mid-May of 2006. Of the samples collected, 29 contained a sufficient amount of water for BC analysis. The rest were analyzed for iron, calcium, and potassium concentration using X-ray fluorescence spectroscopy. These additional aerosol species helped identify possible source regions for the aerosols, including the BC, found in the precipitation.

2.0 Methods

Snow samples were collected in the Sierra Nevada/Cascade mountains and rain samples were collected on the northern California coast from February through May 2006. The precipitation samples were filtered and analyzed for BC mass concentration using a modified version of thermal optical analysis (TOA). Laboratory evaluations were conducted prior to analysis to quantify losses of BC to surface materials and through inefficient filtration. Lastly, smaller precipitation samples were filtered and, using X-ray fluorescence spectroscopy (XRF), analyzed for iron (Fe), calcium (Ca), and potassium (K).
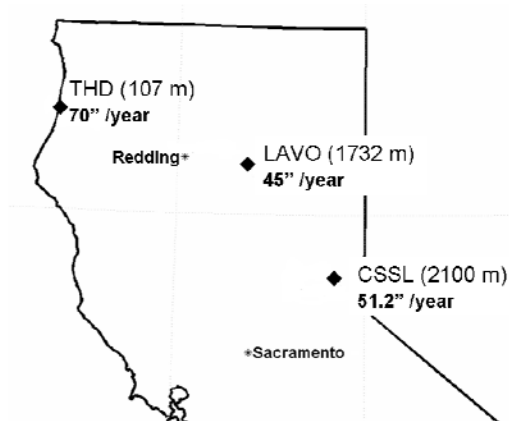
Figure 1. Map of sample collection sites. The elevation is shown in parentheses and average annual precipitation is shown in bold.

2.1. Sample Collection

Three EcoTech automated rain collectors were installed at three separate sites in northern California (Figure 1): two inland mountain sites and one coastal site. Site choices were based both on the scientific relevance of the location as well as installation logistics, i.e., accessibility, available power, and availability of meteorological and other ambient aerosol data.

The Climate Monitoring and Diagnostics Lab station at Trinidad Head (THD), the coastal site, was an ideal choice for collecting precipitation samples from clouds with limited North American continental influence. The site is located on a small point sticking out from the mainland. The largest urban source of pollution is from Eureka, south of Trinidad, and prevailing winter storms generally arrive at THD from the northwest, although the cyclonic rotation of winds under low-pressure systems may pick up some continental pollution.

For snow collection, two sites were chosen, one at the southern tip of the Cascades and the other in the Sierra Nevada. The Interagency Monitoring of Protected Visual Environments (IMPROVE) site at Mt. Lassen Volcanic National Park (LAVO) represented a relatively pristine inland site downwind of Trinidad Head, important for comparing the precipitation from the

same systems as they move across the mountains. Meteorological data as well as aerosol data from the IMPROVE monitoring network were also available here. Additionally, an aethalometer, used to measure ambient BC concentration, and a nephelometer, which measures light scatter as a proxy for total ambient aerosol concentration, were installed at this site. Lastly, the University of California at Davis (UC Davis) installed three-hour-interval continuous aerosol analyzers for the duration of the SUPRECIP-2 study (Suppression of Precipitation Experiment), of which this work was a part.

In contrast to LAVO, the third site, at UC Berkeley's Central Sierra Snow Lab (CSSL), was chosen as a mountain snow collection site likely to be dominated by regional pollution from both San Francisco and Sacramento urban centers and local pollutants from nearby Interstate 80, residences, and trains on the nearby Southern Pacific rail line. Again, meteorological data, as well as aerosol measurements made by the UC Davis team, were available here.

The collectors operated for up to one week without needing visitation (Figure 2). A detector sensed when a precipitation event began, and signaled the collector to open and begin sampling. When the event ended, the collector lid closed, thus protecting the samples from contamination by dry deposition. Samples were separated by days rather than by event, such that for each day of rain or snow, one sample was collected. An overflow valve protected the samples in the event of extreme precipitation events. Each sampler held eight sample bottles with a 400 mL capacity. A computer data logger tracked the date, time, and intensity of each event. Two of the collectors installed at Central Sierra Snow Lab and Lassen Volcanic National Park were modified for snow collection. Heated funnels melted the snow as it fell on the surface and channeled the melt water into a sample bottle. The temperature inside these collectors was held constant at 10°C. Samples from all three sites were collected weekly and frozen until analysis could be performed. Prior to analyzing the precipitation samples for BC, the analysis method was evaluated in the laboratory.

**Figure 2. Rain and snow collection
at Mt. Lassen Volcanic National Park**

Photo Credit: Scripps Institution of
Oceanography

2.2. Laboratory Evaluation

Laboratory standards of known masses of BC suspended in water were used to evaluate
filtration efficiency and surface losses. BC particles were generated with an inverted diffusion
flame of methane and air. This flame produced particles containing mostly BC and a little
organic carbon (*Kirchstetter and Novakov*, 2007). The particles were collected on a stretched
Teflon membrane filter (Gelman, Teflo membrane, 2 μm pore size) and then oxidized by
sampling air with ozone concentrations in excess of 2.9×10^4 ppmv through the loaded filter for
15 minutes. Oxidation transformed the particles from hydrophobic to hydrophilic, crudely
mimicking the atmospheric chemical processing of soot that makes it hydrophilic (*Chughtai, et
al.*, 1996; *Zuberi, et al.*, 2005). After ozone exposure, the particles were exposed to a Polonium-210
Staticmaster Ionizer (model 2U500) to remove static charge, scraped from the filter onto
aluminum foil, and weighed with a Cahn 21 microbalance. Homogeneous colloidal suspensions
of BC particles in water were prepared by sonicating 0.5 to 1.0 mg of the oxidized BC in 100 mL
of distilled water.

The BC laboratory standards were diluted to concentrations of 11.45–45.8 μg L^{-1}, consistent with
previous observations of BC concentrations in precipitation (*Chylek, et al.*, 1999; *Clarke and Noone*,
1985; *Ducret and Cachier*, 1992). Each solution was pulled through a series of three 13-mm-

diameter quartz filters (Pallflex Tissuequartz 2500 QAT-UP) by application of a vacuum, followed by a methanol rinse. The methanol removed water and some organic residues from the loaded filter without affecting the mass of BC retained on the filter. Average BC mass recovery was 92% (±7%), with a slight decrease as filter loading reached maximum values. At the highest loading (9 μg), 90% total recovery was observed. On average, the amount of flame-generated BC lost to surfaces or not captured by the filters was less than 8%.

2.3. Sample Analysis

The version of thermal optical analysis (TOA) used in this study included broadband optical characterization of samples as they were heated. Transmission from 400–900 nm wavelengths was measured with a white-light-emitting diode and a spectrometer. This is the first use of spectral transmission data in estimating the BC content of samples and is described below.

2.3.1. TOA Using Spectral Transmission Data

Total carbon (TC) mass in a sample was computed directly by summing the concentrations of evolved carbon atoms (measured as ppm CO_2 in oxygen at sampling intervals of 1°C over the analysis range 50°C to 700°C) and multiplying by the amount of oxygen in which the carbon atoms were diluted, calculated from the oxygen carrier gas flow rate (0.2 L min^{-1}) and the sample temperature ramp rate (40° min^{-1}):

$$TC = \sum_{T=50°C}^{T=700°C} \frac{\left(\frac{\mu moles\, CO_2}{moles\, O_2} \right)\left(\frac{12\, g\ C}{mol\ C} \right) * \left(\frac{.0409\, mol\ O_2}{L\ O_2} \right) * \left(\frac{0.2 L\ O_2}{min} \right)}{\left(\frac{40°C}{min} \right)}$$

Equation 1. Total carbon determination

Plotting TC as a function of temperature shows that different forms of carbon evolve at different temperatures. Prior work suggests that most non-light-absorbing carbon is organic and evolves between 50°C and 400°C, while light-absorbing carbon generally evolves at temperatures between 450°C and 700°C); however, some initially non-light absorbing organic carbon undergoes pyrolysis during the heating process and may co-evolve with the original BC from the sample (*Mikhailov et al.*, 2006; *Novakov and Corrigan*, 1995) (Figure 3).
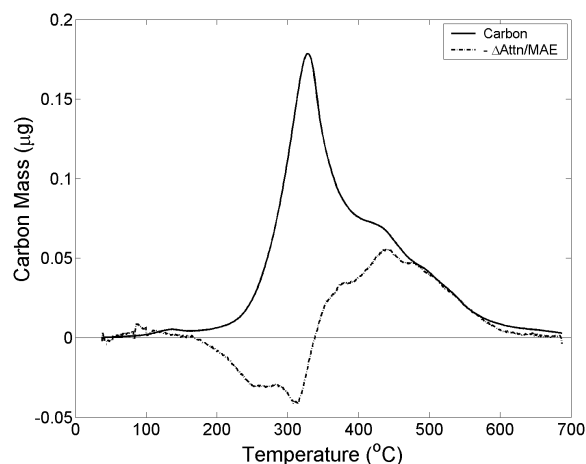
Figure 3. Thermogram of evolved carbon and corresponding transmission (negative attenuation) changes. Change in attenuation is scaled to match carbon mass. Charring of organic carbon obscures the original BC signal.

Source: Scripps Institution of Oceanography

As carbon evolves, the attenuation of light through the filter is measured for wavelengths between 400 and 900 nm. Increases in light attenuation indicate that previously non-light-absorbing aerosols are pyrolizing as temperatures increase. An attenuation decrease corresponds to the oxidation and evolution of light-absorbing carbon. The change in attenuation as a function of temperature can be scaled to match the direct carbon measurement (Figure 3). The scaling factor is called the mass attenuation efficiency (MAE) and can be used to convert change in attenuation to an equivalent BC mass. When significant charring is present, however, this simple conversion no longer applies. Charred organics typically have a much lower MAE than atmospheric BC, so scaling both the BC and charred carbon by the same value will overestimate the amount of BC in the sample. Char may be corrected for using the difference in the spectral dependence of absorption between BC and char.

2.3.2. Modifying the TOA Method for Field Sample Analysis

The absorption spectral dependence of BC was measured using the spectral transmission through laboratory-loaded filters relative to blanks for wavelengths between 500 and 600 nm. The absorption spectral dependence is related to the wavelength of measured light, λ, as λ^{-k}, where k is the absorption angstrom exponent or AAE. The AAE of laboratory BC was 0.75 ± 0.07. The AAE of char was determined using spectral transmission measurements through organic char on backup sample filters when no BC was present and had a value of 5 ± 1. The organic loading on the backup filters was significantly less than what was found on the primary filter and likely due to dissolved organics in the sample adsorbing on the backup filters. The AAE for the BC plus char was calculated at all temperatures between 150°C and 700°C using spectral transmission measurements throughout thermal processing (Figure 4).
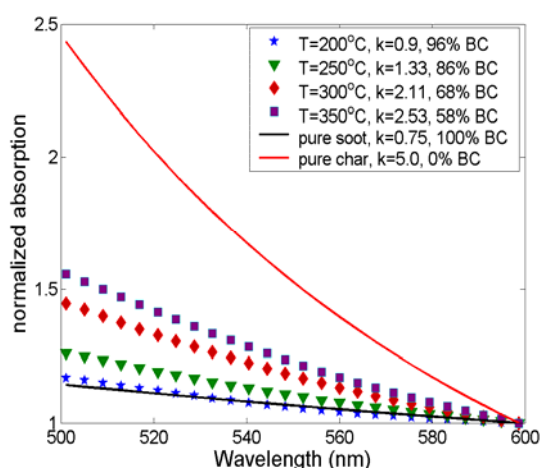
Figure 4. Spectral dependence of light absorption by soot and by char. The angstrom exponent, k, describes change in absorption as a function of wavelength (abs = λ^{-k}).
Source: Scripps Institution of Oceanography

The fraction of attenuation due to each component was scaled by their individual MAE values and added together as the total mass of light-absorbing carbon. An iterative algorithm was used to find the MAE values for both BC and char that provide the best fit to the carbon mass remaining on the filter (derived from direct measurements of thermally evolved CO_2) at temperatures higher than 480°C. (Figure 5). The averaged BC mass retrieval between 150°C and 300°C produced a final value for the original BC in each precipitation sample. This temperature range was used as the central values where the transmission measurements were stable and BC

had not yet begun to evolve. Error estimates were calculated from uncertainties in the derived AAE values and the misfit between the BC and char curves and the direct carbon measurement. For a BC mass loading on the filter greater than 1.5 micrograms, the uncertainty approached 20%. The lower detection limit was approximately 0.4 micrograms.
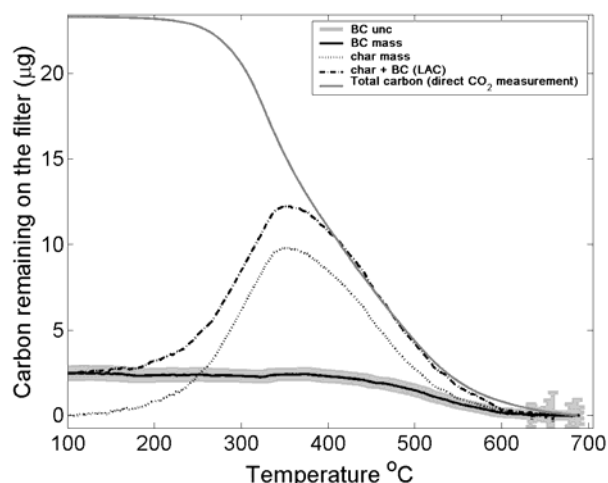
Figure 5. Total carbon remaining on the filter and relative BC and char contribution to total light-absorbing carbon. Total carbon was derived from the direct measurement of evolved CO_2 as a function of temperature.

Source: Scripps Institution of Oceanography

2.4. Elemental Analysis

X-ray fluorescence spectroscopy (XRF) was used to measure the mass concentration of 27 elements in the precipitation samples. Samples were filtered through Watcom Nuclepore (0.4 micron pore size) filters. The amount filtered ranged from 20 to 40 mL. The filters were mounted and subjected to XRF spectroscopy (*VanCuren et al.,* 2005), using the Advanced Light Source Facility (ALS) at Lawrence Berkeley National Laboratory. The beam size is 500 x 500 microns, and five points were measured on each filter to determine uniformity of coverage. Blanks using filtered distilled water gave clean results for Fe, Ca, and K, the elements used in this analysis.

3.0   Results and Discussion

Due to the BC mass loading necessary to make a valid measurement (> 0.4 micrograms), each sample had to accrue at least 100 mL and preferably 200 mL of precipitation. A total of 55 precipitation samples were collected during this field experiment, 29 of which were of sufficient volume to make a BC measurement. Nearly half of these were at THD, where the average BC concentration in rainwater per day was determined for 14 different days. At LAVO, 7 samples were analyzed for BC, and 8 samples were used for BC analysis at CSSL. The other smaller samples at CSSL and LAVO were analyzed for Fe, Ca, and K concentrations and compared with the ambient atmospheric concentration data available for these sites. UC Davis installed aerosol collectors for the duration of the experiment and used XRF to determine the mass concentration of 27 different elements, including Fe, Ca, and K. Combined with air mass back trajectories, these elemental species yield valuable clues to the sources of BC in California precipitation.

3.1.   BC Concentrations in Precipitation

The average BC concentrations in rain at THD, and in snow at CSSL and LAVO, were respectively 5.7 ng/g, 6.9 ng/g, and 5.0 ng/g (Figure 6). Although the difference in average BC concentration is not radically different between the sites, the relative amounts follow what was expected.
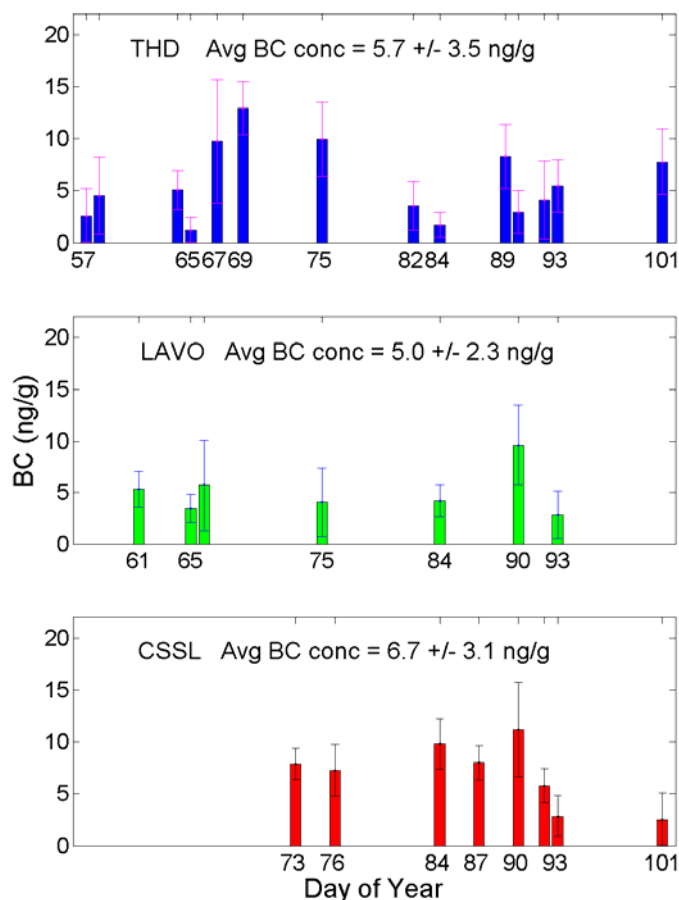
Figure 6. Measured concentrations of BC at THD, LAVO, and CSSL

Source: Scripps Institution of Oceanography

Rainwater at THD, which sits 100 meters above sea level on the northern California coast, contains on average the lowest concentration of BC. THD is the least subject of all the sites to North American influences. However, directly south and less than 30 miles from THD are several major industrial facilities, including saw mills and power plants. Due to typical cyclonic rotation of winds during rain events, which tend to approach THD from the southwest, it is possible that emissions from these facilities may be observed in the rainwater here.

CSSL, at an elevation of 2100 meters, is located directly downwind of Sacramento and San Francisco, as well as next to a small community and ski resort and just a few miles from I-80, a primary trucking route across the Sierra Nevada. CSSL is thus expected to be heavily influenced by these regional and local pollution sources. Close to the source, aerosol pollutants are more concentrated, and therefore concentrations of BC and other aerosols in the precipitation should also be higher due to precipitation scavenging of aerosols from the air below the clouds. Back

trajectories and aerosol analysis, discussed in the following section, provide evidence that this is the case for most storms observed at CSSL.

LAVO, which sits about 60 miles due east of Redding at an elevation of 1732 meters, is both inland and far from major urban or industrial centers. Of the three sites, BC concentration in snow and rain at LAVO more closely represents that found in remote California snowpack and is a combination of regional and long-range transport. Variability found in the BC concentration is much lower than at the other two sites and probably more characteristic of average background concentrations in the Sierra and southern Cascade ranges. Uncertainties measured at LAVO are also higher than at the other two sites. This is because the volumes of water used for the BC analysis were about half that collected at THD and CSSL; thus the BC mass retrieved from these smaller volumes of water tended to be low and therefore subject to higher uncertainty.

The average BC concentration measured at all three sites may be biased low due to preferential sampling of rain and snow from only the heaviest events. This is a result of the aforementioned large volume of sample needed to make a BC measurement, which may result in a dilution of BC concentration. The effects of daily precipitation amount and ambient aerosol concentration on BC and other aerosol species measured in snow and rain are discussed below.

3.2.   Factors Controlling BC Concentration in Precipitation

There are two primary variables controlling the concentration of BC (or any aerosol species) in precipitation. First, the BC concentration in the aerosol particles that participate in the initial formation of cloud droplets (cloud condensation nuclei or CCN), as well as the ambient concentration below the cloud, limits the total BC mass that can be incorporated into the snow or rain. BC in the CCN is subject to "rain out," while particles below the cloud may be scavenged, or "washed out" by falling snow or rain. Depending on the meteorology, the BC particles in the CCN and those below the cloud may or may not be the same. The second variable affecting BC concentration in rain or snow is the amount of precipitable water diluting the concentration, i.e., a heavy rain or snowfall would result in a lower concentration for the same amount of BC. Determining which effect dominates is challenging, as the two variables are not independent of each other. This is clearly demonstrated by Figure 7, which shows that ambient concentrations for all aerosol species tend to be lower during intense precipitation events than for lighter events. This is not surprising, as precipitation is the primary method for clearing the atmosphere of aerosols, i.e., the more rain or snow, the cleaner the air.
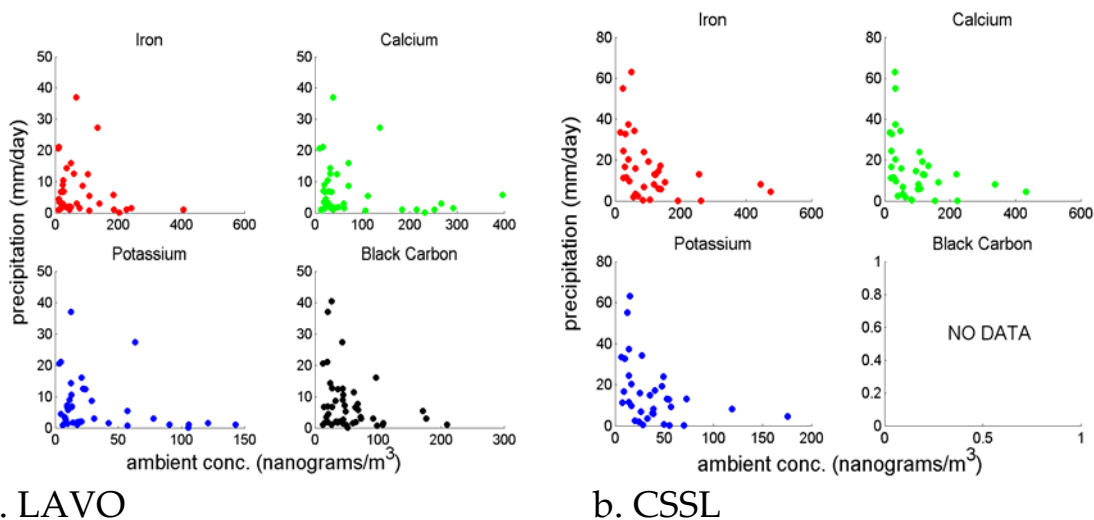
Figure 7. Precipitation daily amount vs. ambient concentrations at (a) LAVO and (b) CSSL. Points are only plotted for days during which precipitation was detected. Due to limited instrument availability, ambient BC data were collected only for LAVO, not CSSL.

Source: Scripps Institution of Oceanography

In the mountains precipitation can form from two different mechanisms: forced orographic lift and synoptic-scale disturbances. Air masses that reach the mountains are pushed upward over the mountains, water condenses during the lifting process, and if there is enough moisture, rain or snow will form. In this type of event, the aerosols at the surface are more likely to represent those in the CCN as well as those that are washed out below the cloud. This type of precipitation event is dominant at CSSL, where total precipitation during this experiment was significantly higher, 737.5 mm, than at LAVO, which equaled 485.68 mm. Precipitation at Trinidad Head during March and April totaled 535.9 mm. For synoptic-scale events, the aerosols measured at the surface may not represent the aerosols which initially fed the clouds. These storm systems are more likely to be influenced by long-range transport of aerosols. During these events, the aerosols observed at the surface may not necessarily represent those in the precipitation, provided that the "rain out" of aerosols dominates over "wash out," which would only occur in remote, pristine areas. Comparison of the Ca, Fe, and K mass concentrations observed in the rainwater to those measured at the surface, as well as a comparison with event intensities, provides insight into the dominating processes.

3.3.   Ambient Aerosol Analysis

LAVO and CSSL precipitation samples smaller than 75 mL were filtered through 0.45 micron Nuclepore filters and analyzed for Ca, Fe, and K using XRF. XRF analysis on rainwater at THD was not done as there were no ambient aerosol measurements there to compare with. The ambient aerosol concentrations at CSSL are highly correlated to those measured in the precipitation; however, a similar comparison at LAVO shows little to no correlation (Figure 8, both red and blue combined). An analysis of three-day air mass back trajectories, calculated using the NOAA (National Oceanic and Atmospheric Administration) HySPLIT model (*Draxler and Hess*, 1998), provides insight into this difference.
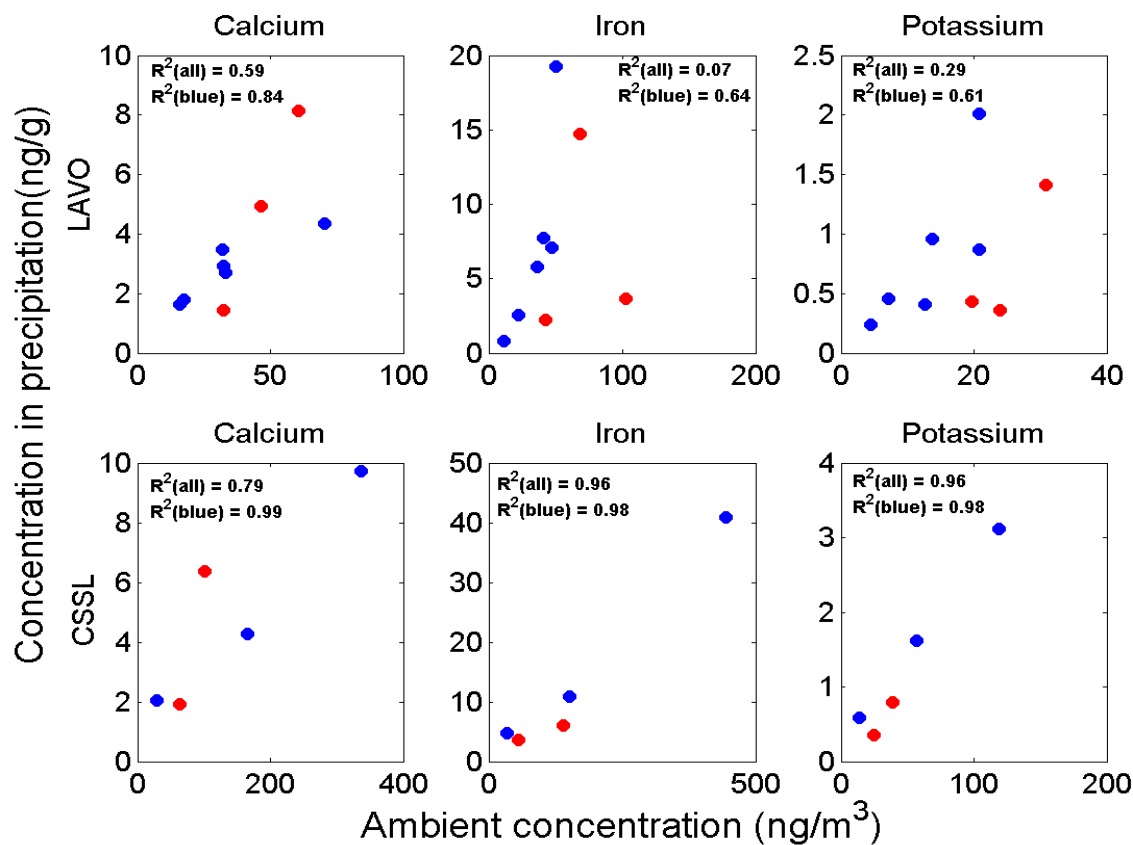
Figure 8. Concentrations of Ca, Fe, and K in precipitation vs. ambient concentrations measured at the surface. Blue points correspond to lower-elevation three-day trajectories (Figure 9), while red points represent days when the trajectories came from altitudes higher than 3.5 km.

Source: Scripps Institution of Oceanography
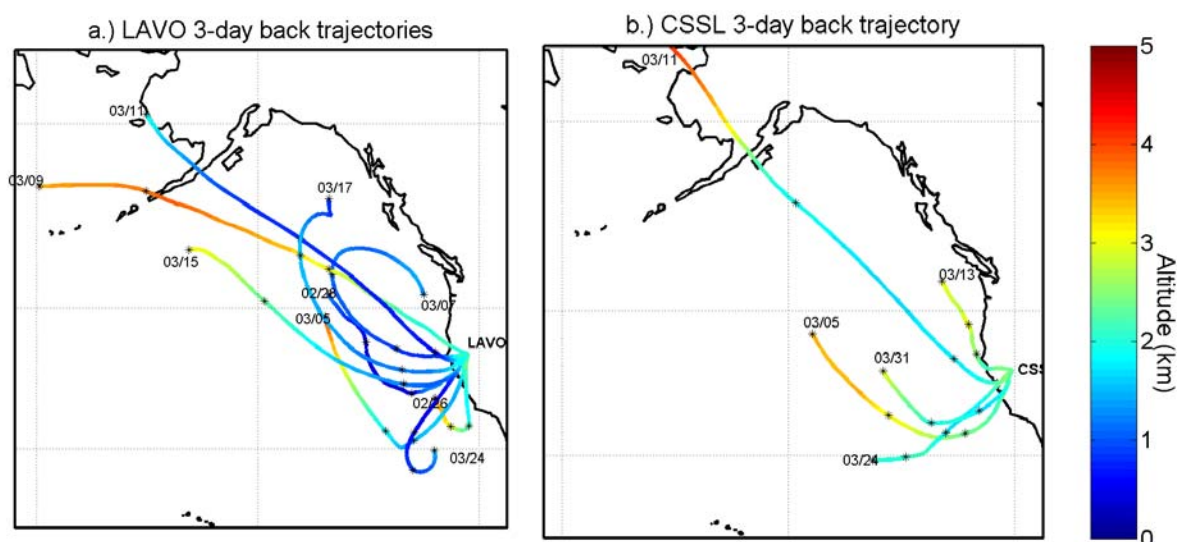
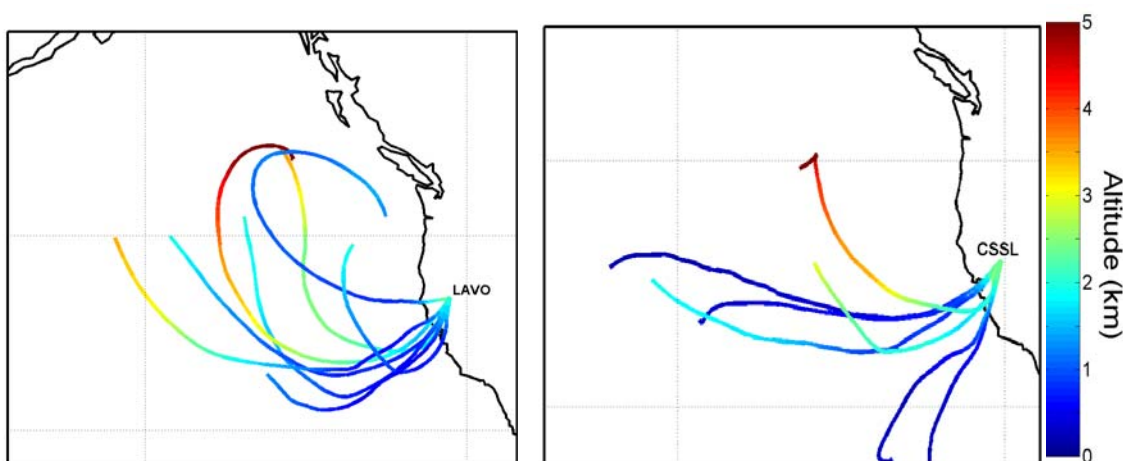**Figure 9. Three-day back trajectories from HySPLIT corresponding to XRF precipitation sample days from (a) LAVO and (b) CSSL. The starting elevation at both sites is 2.4 km, approximately 700 meters above the surface at LAVO and 300 meters above the surface at CSSL**.

Source: Scripps Institution of Oceanography

The XRF rain data are split into two categories based on back-trajectory analysis: air masses arriving from high elevations (<3.5 km) and those arriving from lower elevations (< site elevation) (Figure 9). The correlation between surface ambient aerosols and precipitation concentration increases dramatically at LAVO when only the days corresponding to low-elevation trajectories are used (Figure 8, blue points). Correlations also increase at CSSL; however because there are so few points and the correlation for all three species was originally high, this change has less significance. Furthermore, all trajectories at CSSL show rising motion over the continent in the day prior to the precipitation event and were therefore subject to influence from regional and local pollutants. The low-elevation trajectories indicate convection and a strong likelihood that locally and regionally emitted pollutants are not only well mixed within the boundary layer, but are also the same aerosols feeding the cloud systems. The high correlation between ambient aerosol concentration and that in the precipitation during these days strongly supports this hypothesis.


Figure 10. HySPLIT three-day back trajectories for precipitation events at LAVO and CSSL for which BC measurements were made. The starting elevation at both sites is 2.4 km, approximately 700 meters above the surface at LAVO and 300 meters above the surface at CSSL.

Source: Scripps Institution of Oceanography

The back trajectories corresponding to the events for which BC concentration measurements were made at both LAVO and CSSL are similar to those shown in
Figure 9; however, at CSSL, stronger low-level convection is indicated based on the very low elevations from which the air masses originate (Figure 10). This would explain the heavier precipitation events supplying sufficient sample volumes for BC analysis. A comparison of BC concentration in precipitation to ambient BC concentration at LAVO on the same day shows no correlation; however analysis shows that ambient concentrations of BC just prior to the precipitation event are highly correlated (R^2=0.66) to the total amount of BC deposited during the event (Figure 11). This indicates that below-cloud scavenging is the dominant source of BC in snow at LAVO. Assuming a well-mixed boundary layer (i.e., constant BC concentration) to 2 km above the surface, a comparison between the total BC deposited and the BC in the boundary

layer prior to the event reveals that nearly all the BC in the air is removed by the snow. This is also shown by the drop in ambient BC concentration during the precipitation event to levels near or below the aethalometer's lower limit of detection, roughly 15 ng/m^3. This analysis conserves BC mass and dilution effects need not be considered. Although ambient BC concentration data at CSSL were unavailable for a complete analysis, the average total BC deposition to the snowpack at CSSL was more than a factor of two greater than at LAVO. Thus the differences in BC concentration in snow measured at the "clean" remote site at LAVO and the polluted site downwind of urban and industrial areas at CSSL were not as large as expected due to dilution by heavier precipitation at CSSL.
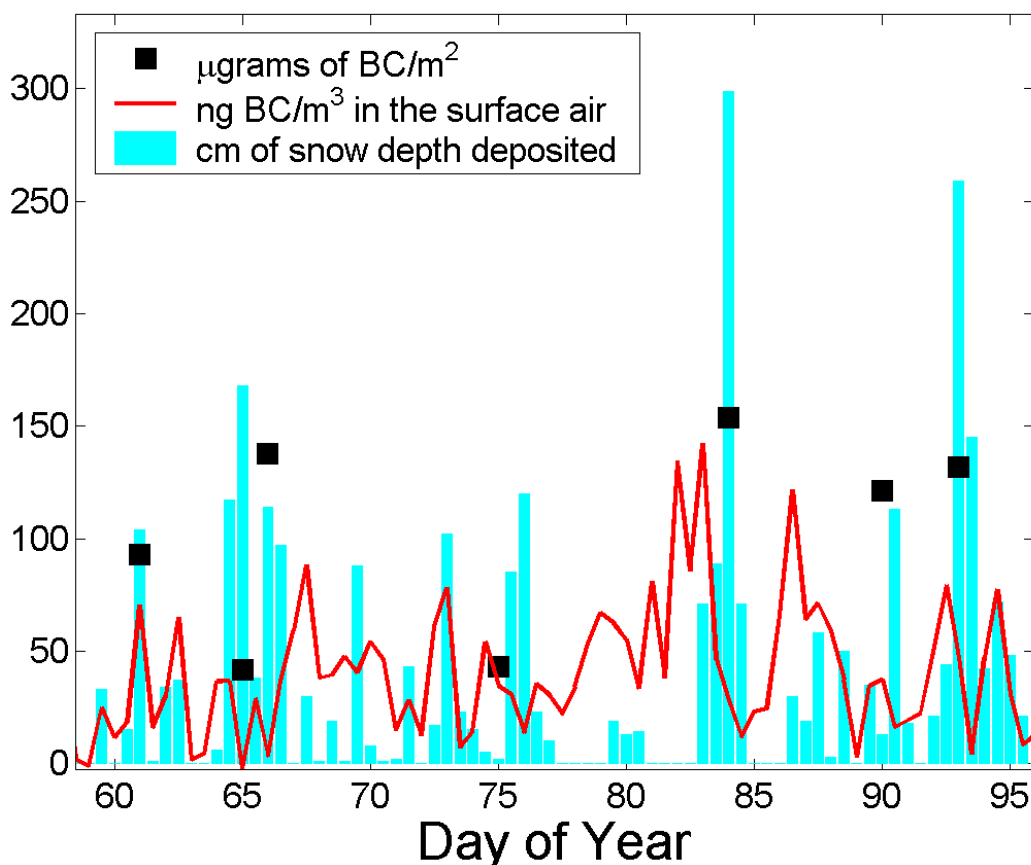
Figure 11. Data are for LAVO. The red line is the ambient concentration of BC/m^3 in the surface air in 12-hour time steps. Measurements were made with a Magee aethalometer. The light blue bars correspond to mm of snow depth accumulated in 12-hour increments. The dark blue squares represent total BC/m^2 deposited by the falling snow during a 24-hour period.

Source: Scripps Institution of Oceanography

At the coastal THD site, three-day back trajectories corresponding to the daily averages of BC concentration in rainwater (Figure 12) indicated primarily marine sources for the aerosol in the rain. For all but two of the days analyzed, air masses approached THD from the southwest

following a cyclonic rotation. On March 8 and 10, however, the air masses approached THD from the northwest. Two trajectories, one three-day back trajectory and one five-day back trajectory corresponding to different times during the precipitation events, clearly indicated trans-Pacific transport from Asia. Two of the highest BC mass concentrations measured in the rain at THD—

9.7 ng/g and 12.9ng/g—correspond to these two days; however, the precipitation amount is also lower than average. The degree to which the high BC concentration was due to transported pollutants or less rainwater diluting the concentration is unclear.
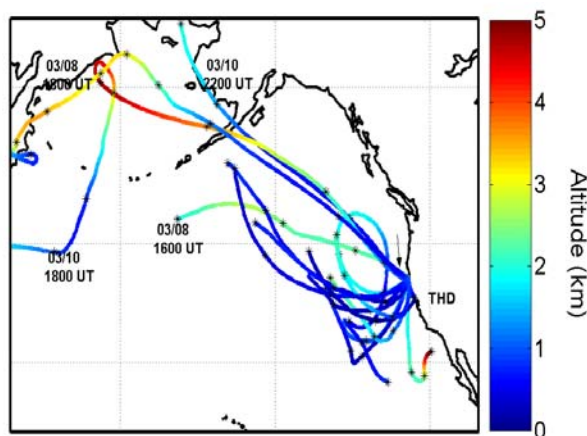
Figure 12. HySplit back trajectories from THD for BC analysis days. Starting elevation at THD is 1 km. The labeled trajectories correspond to those approaching THD from the NW; these days are unique compared to all the other trajectories approaching THD cyclonically from the SW.

Source: Scripps Institution of Oceanography

A comparison of the total precipitation in a given day and the corresponding concentration of aerosol species in the precipitation at the three sites was inconclusive. It is difficult to isolate the precipitation dilution effect from the ambient aerosol influence. No statistically significant trend was observed at either LAVO or CSSL for the XRF aerosol data, although removal of one or two outliers in the LAVO data increased the correlation and revealed the possibility of a trend. A weak negative correlation (R^2=0.3) was observed between BC concentration in the snow and total amount of snow accumulation at LAVO. Removal of four of the 14 points at THD resulted in a strong negative correlation between the rain amount and BC concentration.

4.0   Conclusions and Recommendations

The average concentration of BC in rain at Trinidad Head was 5.7 ± 3.5 ng/g of water. The average concentrations of BC in snow at the mountain sites were 5.0 ± 2.3 ng/g at LAVO and 6.9 ± 3.1 ng/g at CSSL. Similar measurements made by Chylek et al (1999) near Halifax, Nova Scotia, showed that BC concentration in snow in rural Nova Scotia averaged 1.7 ± 0.83 ng/g, while in urban snow near Halifax averaged 11 ± 7.7 ng/g. The measured concentrations of BC in California's snowpack are therefore typical of an urban environment, although very few measurements of these kinds exist for comparison. The results from this study are based on six weeks of measurements taken at three sites in California. They suggest that BC has significant impact on the snowpack; however, more complete information about BC effects on the snowpack will require a broader network of observation stations and a longer time series of data.

The average concentration of BC in snow reported in this study may be biased low, as heavier precipitation events may dilute the concentration compared to lighter events. As the albedo of the snow surface depends only on the first few centimeters of snow depth, higher concentrations of BC in light snowfall would initially have just as strong an impact on surface albedo as BC in heavy snowfall.

Aerosol analysis and air-mass back-trajectory calculations for both CSSL and LAVO point to a strong influence from regional and local sources. Surface air concentrations of Fe, Ca, and K are highly correlated to the concentrations measured in the precipitation when back trajectories indicate convection over land prior to arrival at both LAVO and CSSL. On days for which BC measurements in precipitation were collected, the back trajectories generally indicate low-level convection prior to arrival at the site. This is likely due to the direct relationship between the strength and level of convection and precipitation intensity.

Total BC deposition shows strong correlation with ambient BC concentrations just prior to each precipitation event at LAVO, indicating that below-cloud scavenging is a primary source of BC in snow at LAVO and likely at other remote mountain regions. Lack of ambient BC concentration data at CSSL and THD prevented a similar comparison for these sites. Although it appears that most of the aerosol sources for snow contaminants are local, a few of the back trajectories at LAVO reveal air masses originating from altitudes greater than 3.5 km from the west, a clear possibility for transported pollutants. This study did not determine whether the surface aerosols that were compared to the contaminants in the precipitation were regionally generated or the result of trans-Pacific transport; however, previous studies have shown that transported pollutants are often observed at high elevations in the Sierra Nevada (*VanCuren and Cahill,* 2002; *VanCuren et al.,* 2005) and therefore may influence BC concentration in snow. At THD, the two highest observed concentrations correspond to days where air mass back trajectories indicate direct trans-Pacific transport. Back-trajectory calculations for the other sample days show that the air masses spent more time over the ocean and, furthermore, show

strong cyclonic rotation over the Pacific Ocean, representing strong precipitation events that would clear out aerosols prior to arrival at THD.

The results of several modeling studies indicate the BC concentration found in the snow at the mountain sites would lead to a reduction in surface albedo for fresh snow between 0.35% and 1.0% for wavelengths between 400 and 700 nm, assuming snow crystals effective radius of 100 microns (*Flanner et al.*, 2007; *Hansen and Nazarenko*, 2004). This change in albedo would lead to an additional 0.7 to 2.0 W/m^2 absorbed at the surface of the snowpack under clear skies at this location and time of year. In other words, if the ambient temperature were near zero, the extra energy absorbed by the snow due to a 1% decrease in albedo would be enough to melt an additional 21.5 g/m^2 of snow per hour. All model results show that for larger snow crystals and aged snow, the effectiveness of BC at reducing albedo and absorbing solar radiation increases dramatically. This could lead to a strong positive feedback in early springtime snow melt.

Previous studies (*Zhu, et al.*, 2007) have shown that the trans-Pacific transport of Asian pollutants, including a significant amount of transported BC (*Hadley, et al.*, 2007), peaks in mid-April. No snow samples were collected during mid to late April at either LAVO or CSSL, so it is still unclear whether enhanced transport will have a discernable effect on melt rates, or if local and regional pollutants overwhelm the contribution from trans-Pacific transport. Dry deposition of BC during periods of atmospheric subsidence, when high-altitude pollutants are more effectively transported to the surface, may also play a role in reducing snowpack albedo.

A major uncertainty in determining BC's effect on snow albedo and thus the melt rate results from a lack of understanding as to how BC vertically redistributes itself through the snowpack during the melt season. Many models assume that as the snow melts, that BC is left behind in the surface layers further amplifying its effect, although it is possible that much of the BC is flushed through the snow with the melt water. Additional field studies are needed to determine the vertical redistribution of BC throughout the snowpack during the melt season. Direct measurements of snow albedo and the effective radius of the snow crystal size would help validate the relationship between albedo and BC concentration.

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6.0 Glossary

AAE   absorption angstrom exponent

ATN   attenuation: a measure of amount of light passing through the aethalometer's sampling filter, proportional to the logarithm of the amount of transmitted light

BC   black carbon: the primary light-absorbing aerosol species that contributes to global warming and regional climate change

CCN   cloud condensation nuclei

CSSL   Central Sierra Snow Laboratory, located near Donner Summit in California

LAVO   Lassen Volcanic National Park, located east of Redding, California

MAE   mass attenuation efficiency: a measure of how much light is attenuated per unit mass of an aerosol material on a filter; dividing the aerosol absorption coefficient by the MAE gives the mass concentration of the absorbing aerosol

SUPRECIP Suppression of Precipitation Experiment, a research effort to study how aerosol pollutants affect cloud formation

TC   Total carbon

THD   Trinidad Head, located in Trinidad, California, on the coast

TOA   thermal-optical analysis: a method of measuring the carbon content of atmospheric particulate matter collected on quartz filters

UC   University of California

XRF   X-ray fluorescence spectroscopy